

UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE BOARD OF PATENT APPEALS
AND INTERFERENCES

Ex parte GARY G. PODREBARAC,
WILLIBRORD A. GROTON, and LAWRENCE A. SMITH, JR.

Appeal 2008-0118
Application 10/820,399
Technology Center 1700

Decided: October 31, 2007

Before BRADLEY R. GARRIS, CHARLES F. WARREN, and
THOMAS A. WALTZ, *Administrative Patent Judges*.

WARREN, *Administrative Patent Judge*.

DECISION ON APPEAL

Applicants appeal to the Board from the decision of the Primary Examiner finally rejecting claim 13 in the Office Action mailed August 2, 2005. 35 U.S.C. §§ 6 and 134(a) (2002); 37 C.F.R. § 41.31(a) (2006).

We affirm the decision of the Primary Examiner.

Claim 13 illustrates Appellants' invention of a process for removal of sulfur from a full boiling range fluid cracked naphtha stream:

13. A process for removal of sulfur from a full boiling range fluid cracked naphtha stream comprising the steps of:

- (a) feeding hydrogen and a full boiling range naphtha feed containing olefins, diolefins, mercaptans, thiophene and other organic sulfur compounds to a distillation column reactor;
- (b) concurrently in said distillation column reactor:
 - (i) reacting a portion of the mercaptans contained within said full boiling range naphtha stream with a portion of the diolefins contained within said full boiling range naphtha stream to produce sulfides and
 - (ii) separating said full boiling range naphtha stream into three fractions by fractional distillation;
- (c) removing product from said distillation column reactor comprising a light naphtha containing substantially no mercaptans, sulfides or other organic sulfur compounds as an overheads;
- (d) removing an intermediate naphtha as a side draw from said distillation column reactor containing thiophene, diolefins boiling in the range of thiophene and mercaptans boiling in the range of thiophene;
- (e) removing a heavy naphtha from said distillation column reactor containing said sulfides and other organic sulfur compounds as a bottoms;
- (f) feeding said intermediate naphtha to a single pass fixed bed reactor containing a hydrodesulfurization catalyst where substantially all of any remaining sulfides and other organic sulfur compounds are reacting with hydrogen to form hydrogen sulfide.

The Examiner relies upon the evidence in these references (Answer 2):

Hearn	5,597,476	Jan. 28, 1997
McDaniel	US 2003/0136706 A1	Jul. 24, 2003

Appellants request review of the ground of rejection under 35 U.S.C. § 103(a) advanced on appeal: claim 13 as being unpatentable over Hearn in view of McDaniel. Ans. 3; Br. 10.

The issue in this appeal is whether the Examiner has carried the burden of establishing a *prima facie* case in the ground of rejection advanced on appeal.

The plain language of claim 13 specifies a process comprising at least the steps of first, feeding any amount of hydrogen and any amount of a full boiling range fluid cracked naphtha feed containing any amount of at least olefins, diolefins, mercaptans, thiophene and other organic sulfur compounds to any manner of distillation column reactor. Second, in the distillation column reactor, concurrently reacting a portion, however small, of the mercaptans in the feed with a portion, however small, of the diolefins in the feed to produce disulfides, and separating the thus treated feed into three fractions which are moved from the column. The three removed fractions are: any amount of an overheads product comprising at least any amount of a light naphtha containing substantially no mercaptans, sulfides or “other sulfur compounds” such as thiophenic compounds; any amount of a side draw intermediate naphtha containing at least any amount, however small, of thiophene, diolefins boiling in the range of thiophene, and mercaptans boiling in the range of thiophene; and any amount of a bottoms heavy naphtha containing any amount of sulfides and other organic sulfur compounds. There is no limitation on the three fractions other than any amount, however small, of the stated components, including impurities. In this respect, the products overhead can contain in-substantial amounts of “other organic compounds,” such as thiophene and other thiophenic compounds. And, third, feeding the side-draw intermediate naphtha fraction to any manner of a single pass fixed bed reactor containing any amount of any manner of hydrodesulfurization catalyst to react “substantially all of any

remaining sulfides and other organic sulfur compounds,” including thiophene, diolefins boiling in the range of thiophene, and mercaptans boiling in the range of thiophene, with hydrogen to form hydrogen sulfide, without limitation on the hydrodesulfurization process parameters. The hydrodesulfurization catalysts can “include Group VIII metals, such as cobalt, nickel, palladium, alone or in combination with other metals such as molybdenum or tungsten on a suitable support which may be,” among other things, alumina. *See* Specification 7:6-11.

The open-ended terms “comprising” and “containing” open the claim to include processes having any manner of other steps and any manner of any amount of additional catalysts, reactants, reagents, and impurities. *See, e.g., Exxon Chem. Pats., Inc. v. Lubrizol Corp.*, 64 F.3d 1553, 1555 (Fed. Cir. 1995) (“The claimed composition is defined as comprising - meaning containing at least - five specific ingredients.”); *In re Baxter*, 656 F.2d 679, 686 (CCPA 1981) (“As long as one of the monomers in the reaction is propylene, any other monomer may be present, because the term ‘comprises’ permits the *inclusion* of other steps, elements, or materials.”). For Example, the side draw intermediate naphtha fraction can contain thiophenic compounds other than thiophene, such as 2- and 3-methylthiophene, as well as benzothiophenic compounds, and indeed, any manner of “any remaining sulfides” as specified in clause (f) to be in this fraction. *See* Specification, e.g., 1:9-12, 4:7-10, and 12:33-36 and 38.

We find Hearn would have disclosed to one of ordinary skill in this art a desulfurization process for olefin- and diolefin-containing, full boiling range fluid cracked naphtha feed stream to remove mercaptans, thiophene, hydrogen sulfide, and other sulfur containing compounds by reacting

mercaptans with diolefins to produce disulfides in a first reactive distillation column, and removing the sulfur compounds by destructive hydrogenation in a second reactive distillation column. Hearn, e.g., Abstract, col. 1, ll. 8-17, col. 2, ll. 10-12 and 30-63, col. 3, ll. 17-64, col. 4, l. 9 to col. 6, l. 14, col. 6, l. 44 to col. 7, l. 28, and Fig. The process stream in the first reactive distillation column is separated into two fractions. The first fraction is a distillate product overheads fraction having reduced mercaptan content which contains most of the olefins and is not subjected to harsh hydrogenation conditions of the bottoms fraction. The second fraction is a bottoms fraction of heavy naphtha containing sulfides, thiophenes and heavier mercaptans which, along with hydrogen, is fed to the second reactive distillation column containing a hydrodesulfurization catalyst to hydrotreat the bottoms by destructively hydrogenating the sulfur containing compounds to form hydrogen sulfide which is removed by fractionation. *Id.*, e.g., Abstract, col. 1, ll. 13-17, col. 2, ll. 44-62, col. 3, ll. 34-64, col. 4, l. 1 to col. 5, l. 15, col. 5, l. 63 to col. 6, l. 14, and col. 7, ll. 6-25. Hearn discloses that “[w]ithout the concern for the olefins, and the necessity to carry out a destructive hydrogenation of the sulfides and other sulfur compounds” in the second reactive distillation column, there can be used a “hydrodesulfurization catalyst comprising two metal oxides supported on an alumina base, said metal oxides chosen from the group consisting of molybdenum, cobalt, nickel, tungsten and mixtures thereof are preferred.” *Id.*, e.g., col. 4, l. 64 to col. 5, l. 3, and col. 5, l. 63, to col. 6, l. 14. Hearn discloses that “[t]he sulfur content of the catalytically cracked fractions will depend upon the sulfur content of the feed to the cracker as well as the

boiling range of the selected fraction used as feed to the process.” *Id.* col. 4, ll. 19-25.

We find McDaniel would have disclosed to one of ordinary skill in this art a desulfurization process for olefin-containing, full boiling range fluid cracked naphtha feed stream to remove sulfur-containing impurities, including mercaptans, thiophenic compounds, benzothiophenic compounds, and organic sulfides, by first treating at least part of the thiophenic compounds in the feed stream with an olefin-modification catalyst in a reactive distillation column to form, among other things, “refractive” thiophenic compound that “can only be treated by conventional hydrodesulfurization means which results in undesirable concomitant octane loss.” McDaniel, e.g., Abstract, ¶¶ 0016, 0020, 0023, 0053, 0060, 0086-0089, and 0103-0109, and the Fig. McDaniel discloses “refractive sulfur is a thiophene containing seven or more alkyl carbons.”¹ *Id.* ¶ 0089. The olefin-modification catalyst mediates the alkylation of at least part of the thiophenic compounds with the olefins in the feed stream to form higher boiling thiophenic derivatives, including “refractive” sulfur derivatives, and reduce the amount of olefins in the stream. *Id.*, e.g., ¶¶ 0026, 0034, 0049, 0061, 0068-0089, and 0105, and the Fig. The thiophenic compounds in the feed stream can include thiophene, 2-methyl- and 3-methylthiophene, 2-ethyl- and 3-ethylthiophene, and 2,3-dimethyl and 2,5-dimethylthiophene. *Id.* ¶¶ 0060, 0078, and 0079.

¹ We find one of ordinary skill in the art would have determined from McDaniel as a whole that the term “refractive” is used in the sense of “resistant to treatment” which is a definition of the term “refractory.” See, e.g., **refraction and refractory**, *The American Heritage Dictionary of The English Language* 1468 (4th ed., Boston, Houghton Mifflin Company, 2000).

McDaniel discloses the resulting olefin-modification product stream is separated into at least three fractions, with the “advantage . . . that the refractive compounds . . . can be recovered in a relatively small volume stream of the highest boiling fraction” taken off as bottoms. The other fractions include the lowest boiling fraction which is a “relatively desulfurized,” that is, “sulfur-lean,” gasoline product taken off as overheads; and the side-draw intermediate boiling fraction which is hydrotreated in a manner wherein the octane number is retained while the sulfur-containing compounds are converted to hydrogen sulfide. McDaniel, e.g., Abstract, ¶¶ 0016, 0017, 0023-0040, 0047-0051, 0090-0109, 0093-0095, and 0100-0109, and the Fig. McDaniel contrasts this process with an analogous prior art process in which the olefin-modification product is divided into two fractions and a hydrodesulfurization reaction carried out with the lower boiling fraction results in undesirable octane loss in the product fraction because of said reaction. *Id.* ¶¶ 0021 and 0015.

McDaniel discloses the sulfur-containing impurities in the side draw intermediate fraction are converted at least in part to hydrogen sulfide by one of three hydrotreating processes: (1) contact with a selective hydrotreating catalyst; (2) “operating a conventional hydrotreating process at selective hydrotreating conditions that are relatively less severe such that the desulfurization occurs while limiting olefin saturation;” and (3) using “a hydrodesulfurization zone followed by a reforming zone to increase the octane number.” McDaniel, e.g., ¶¶ 0031, 0039, 0041, 0050, 0094, and 0100-0102. The hydrodesulfurization catalyst can include conventional catalysts, such as a Group VI and/or Group VIII metal on a support, wherein

the typical metals are molybdenum, tungsten, nickel and cobalt, and the support is typically alumina. *Id.* ¶ 0096.

McDaniel discloses the side draw intermediate boiling fraction can be in the range of less than about 120°C to less than about 240°C, with the product overheads fraction and the bottom fraction below and above this range, respectively. McDaniel, e.g., ¶¶ 0028-0030 and 0036-0038.

McDaniel discloses the boiling point of thiophene is 84°C and the boiling point of mono- and di- C₁₋₂-alkyl thiophenes would fall within the boiling point range of the side drawn intermediate fraction. *Id.* ¶¶ 0077 and 0128, and Table V. The data in Table V from the process illustrated in Example 2, shows that thiophene is present in the first fraction; tetrahydrothiophene and C₁₋₂ thiophene are present in the first two fractions, and other thiophenic compounds, including “refractive” alkylated thiophenes, are present at least to some extent in each of the three fractions. McDaniel discloses the three fractions shown in Table V are “prepared in accordance with the present invention” even though “the fractionation that was carried out was less than ideal” because the first fraction “IBP-100°C” contains higher boiling thiophene compounds. *Id.* ¶¶ 0123-0128.

We determine the combined teachings of Hearn and McDaniel, the scope of which we determined above, provide convincing evidence supporting the Examiner’s case that the claimed invention encompassed by claim 13, as we interpreted this claim above, would have been *prima facie* obvious to one of ordinary skill in the fluidized catalytic cracking processing arts familiar with processes of removing sulfur from full boiling range fluid cracked naphtha feed streams.

We determine that one of ordinary skill in this art would have combined Hearn and McDaniel because each is directed to processes of removing at least part of the same sulfur impurities from the same naphtha feed stream to produce a product overheads that is substantially free of sulfur impurities. Further, this person would have recognized that each reference reacts olefins with different sulfur impurities using a different catalyst to form different sulfur impurities in a reactive distillation column from which the resulting impurities are removed in a bottoms fraction and subsequently converted to hydrogen sulfide using a hydrodesulfurization catalyst. Thus, this person would have further recognized that the respective processes essentially differ in the manner of processing that part of the process stream that is not removed as product overheads.

We further determine that McDaniel would have reasonably suggested to this person that part of the process stream that is not removed as product overheads can be split into at least two fractions wherein the side draw intermediate fraction can be hydrotreated to reduce sulfur impurities contained in the fraction with processes ranging from to light to severe with respect to the desired effect on the olefin content of the fraction; the severe treatment being a hydrodesulfurization process. This person would have further found in McDaniel the teachings that the boiling point range of the three fractions can be varied depending on the sulfur impurities, and thus, can be selected to maintain the product overheads substantially sulfur impurity free. We note, in this respect, that the sulfur impurities in the naphtha feed stream vary as pointed out by Hearn.

We still further determine that the combination of Hearn and McDaniel would have reasonably suggested to one of ordinary skill in this

art that the second fraction containing the sulfur impurities removed from the reactive distillation column in Hearn's process can be split into at least two parts as suggested by McDaniel, and that based on known boiling point data, the thiophenic compounds, including thiophene, taught by Hearn to be in the second fraction, can be in a side draw middle fraction which can be subjected to a hydrotreating process of severity suitable to the impurities as taught by McDaniel. We determine that one of ordinary skill in this art would have found in McDaniel the teaching that a suitable hydrotreating process includes a conventional hydrotreating process which is less severe than a hydrodesulfurization process even though a hydrodesulfurization catalyst is used in such processes.

Accordingly, we are of the opinion that, *prima facie*, one of ordinary skill in this art routinely following the combined teachings of Hearn and McDaniel would have reasonably arrived at the claimed process for removing sulfur from a full boiling range fluid cracked naphtha stream encompassed by claim 13, including all of the limitations thereof arranged as required therein, without recourse to Appellants' Specification. *See, e.g., KSR Int'l Co. v. Teleflex Inc.*, 127 S. Ct. 1727, 1740 (2007) ("if a technique has been used to improve one device, and a person of ordinary skill in the art would recognize that it would improve similar devices in the same way, using the technique is obvious unless its actual application is beyond his or her skill"); *In re Kahn*, 441 F.3d 977, 985-88 (Fed. Cir. 2006); *In re Dow Chem. Co.*, 837 F.2d 469, 473 (Fed. Cir. 1988);² *In re Keller*, 642 F.2d 413,

² The consistent criterion for determination of obviousness is whether the prior art would have suggested to one of ordinary skill in the art that [the claimed process] should be carried out and would have a reasonable likelihood of success viewed in

425 (CCPA 1981);³ *In re Sovish*, 769 F.2d 738, 743 (Fed. Cir. 1985) (skill is presumed on the part of one of ordinary skill in the art); *In re Bozek*, 416 F.2d 1385, 1390 (CCPA 1969) (“Having established that this knowledge was in the art, the examiner could then properly rely, as put forth by the solicitor, on a conclusion of obviousness ‘from common knowledge and common sense of the person of ordinary skill in the art without any specific hint or suggestion in a particular reference.’”); *see also In re O’Farrell*, 853 F.2d 894, 903-04 (Fed. Cir. 1988) (“For obviousness under § 103, all that is required is a reasonable expectation of success.” (citations omitted)).

Furthermore, one of ordinary skill in this art would have selected a suitable workable or optimum boiling point range for the side drawn intermediate fraction to contain sulfur impurities, including thiophene, which can readily be removed by a suitable hydrotreating process achieving the desired result with respect to olefin content as taught by McDaniel. *See, e.g., In re Boesch*, 617 F.2d 272, 275-76 (CCPA 1980) (the prior art would have suggested the experimentation necessary to achieve the claimed compositions as discovery of an optimum value of a result effective variable in a known process is ordinarily within the skill of the art); *In re Aller*, 220

light of the prior art. [Citations omitted] Both the suggestion and the expectation of success must be founded in the prior art, not in the applicant’s disclosure.

Dow Chem., 837 F.2d at 473.

³ The test for obviousness is not whether the features of a secondary reference may be bodily incorporated into the structure of the primary reference; nor is it that the claimed invention must be expressly suggested in any one or all of the references. Rather, the test is what the combined teachings of the references would have suggested to those of ordinary skill in the art.

Keller, 642 F.2d at 425.

F.2d 454, 456-58 (CCPA 1955). (it is not inventive to discover by routine experimentation optimum or workable ranges for general conditions disclosed in the prior art).

Appellants' contentions do not successfully rebut the *prima facie* case. We cannot agree with Appellants' contention that all of the thiophene and other thiophenic compounds are alkylated to refractive alkyl substituted thiophenes in McDaniel's process. Br. 13-14. Indeed, McDaniel teaches that only a portion of the thiophenic compounds can be converted to refractive thiophenes, and thus, thiophene and other thiophenic compounds can be present in one or more of the three fractions removed from the first reactive distillation column depending on the respective boiling ranges of the fractions.

In this respect, we recognize that the data for the process of McDaniel's Example 2 reported in Table V shows thiophene is present only in the first fraction. However, one of ordinary skill in this art would have expected this result with a first fraction boiling range of 100°C as the boiling point of thiophene is about 84°C as set forth in McDaniel. A first fraction having a lower boiling range would reasonably be expected to result in thiophene present in the first two fractions at least to some extent, which is all that claim 13 requires as we interpreted this claim above. We further cannot agree with Appellants' contention that McDaniel's Table V is deceptive and not in line with the invention disclosed therein. Br. 15. Although McDaniel recognizes that the process conducted is "less than ideal," the result reflects the disclosed invention because "the respective three fractions [are] prepared in accordance with the present invention."

McDaniel ¶ 0128.

Accordingly, based on our consideration of the totality of the record before us, we have weighed the evidence of obviousness found in the combined teachings of Hearn and McDaniel with Appellants' countervailing evidence of and argument for nonobviousness and conclude that the claimed invention encompassed by appealed claim 13 would have been obvious as a matter of law under 35 U.S.C. § 103(a).

The Primary Examiner's decision is affirmed.

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No time period for taking any subsequent action in connection with this appeal may be extended under 37 C.F.R. § 1.136(a)(1)(iv) (2007).

AFFIRMED

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